



Green remediation and recycling of contaminated sediment by waste-incorporated stabilization/solidification



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HIGHLIGHTS

- Green remediation of contaminated sediment with waste materials enables recycling.
- Compressive strength is inversely correlated to average pore diameter.
- Coal fly ash outperforms ground seashells and lime to augment cement hydration.
- Combustion bottom ashes and glass cullet enhance strength for reuse applications.
- Metals are immobilized in the sediment blocks with minimal leachability.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 11 September 2014
 Received in revised form 12 November 2014
 Accepted 30 November 2014
 Available online 15 December 2014

Handling Editor: X. Cao

Keywords:

Contaminated sediment
 Green remediation
 Waste recycling
 Stabilization/solidification

ABSTRACT

Navigational/environmental dredging of contaminated sediment conventionally requires contained marine disposal and continuous monitoring. This study proposed a green remediation approach to treat and recycle the contaminated sediment by means of stabilization/solidification enhanced by the addition of selected solid wastes. With an increasing amount of contaminated sediment (20–70%), the 28-d compressive strength of sediment blocks decreased from greater than 10 MPa to slightly above 1 MPa. For augmenting the cement hydration, coal fly ash was more effective than lime and ground seashells, especially at low sediment content. The microscopic and spectroscopic analyses showed varying amounts of hydration products (primarily calcium hydroxide and calcium silicate hydrate) in the presence of coal fly ash, signifying the influence of pozzolanic reaction. To facilitate the waste utilization, cullet from beverage glass bottles and bottom ashes from coal combustion and waste incineration were found suitable to substitute coarse aggregate at 33% replacement ratio, beyond which the compressive strength decreased accordingly. The mercury intrusion porosimetry analysis indicated that the increase in the total pore area and average pore diameter were linearly correlated with the decrease of compressive strength due to waste replacement. All the sediment blocks complied with the acceptance criteria for reuse in terms of metal leachability. These results suggest that, with an appropriate mixture design, contaminated sediment and waste materials are useful resources for producing non-load-bearing masonry units or fill materials for construction uses.

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1. Introduction

Dredging in harbors is necessary to maintain sufficient depth for navigational access. More than 600 Mm³ of marine sediments are dredged annually throughout the world, which, however, are considered as waste materials with less than 1% being recycled at present (Dubois et al., 2009; Wang et al., 2013). Across the coasts of China, metal contamination of marine sediments has been detected in association with accelerated economic growth in the past decades (Pan and Wang, 2012). In Hong Kong, elevated levels of heavy metals in marine sediments were also found at localized hotspots (e.g., typhoon shelters that were embayment with low flushing capacity due to breakwaters) due to historical pollution by industrial and domestic sewage discharges (HK EPD, 2013). Significant amounts of dredged sediments in Hong Kong were classified as unacceptably contaminated (approximately 29 Mm³) and required disposal at contained marine disposal facilities with clay capping and continuous monitoring programme (HK EWTB, 2002; HK CEDD, 2008), which could present a long-term liability. Although *ex situ* remediation technologies such as chelant-enhanced washing and electrokinetic extraction have been investigated for contaminated sediments (Polettini et al., 2007; Kirkelund et al., 2010), their full-scale applications were limited by substantial cost and time implications. Proper remediation of contaminated sediments is an imperative challenge that presents significant financial and environmental burdens.

Cement-based stabilization/solidification of contaminated sediments has proved to be an appealing technology for metal immobilization and strength improvement, such that the treated sediments can be recycled as aggregate for road construction (Dubois et al., 2009; Pinto et al., 2011). Nevertheless, the contaminated marine sediments interfered with the chemistry of cement hydration, compromising the effectiveness of metal stabilization and development of compressive strength in the cementitious materials (Limeira et al., 2010; Wang et al., 2013). The presence of organic matter enhanced calcium complexation and pH buffering capacity that hindered the cement hydration and pozzolanic reactions (Dubois et al., 2009), whereas soluble chloride and sulfate salts in marine sediments led to undesirable metal mobility (Zentar et al., 2012; Zhao et al., 2013). Moreover, heavy metals resulted in delayed setting and weak early strength via metal doping and isomorphic substitution in the hydrated and crystalline phases (Gollmann et al., 2010; Pandey et al., 2012; Navarro-Blasco et al., 2013). Previous studies investigated pretreatment methods such as water/acid extraction and chemical oxidation for mitigating the detrimental effects of organic matter, soluble salts, and heavy metals (Zentar et al., 2009; Colangelo et al., 2012). However, additional wastewater treatment processes, intensive consumption of energy and chemicals, as well as inevitable increase in treatment costs and duration still remain as the major barriers to widespread applications. As a result, the potential recycling of contaminated sediments has so far been limited to the controlled utilization as low-strength materials in road subgrade and base formation in many cases (Naganathan et al., 2013; Zhen et al., 2013).

A new remediation strategy that utilizes agricultural/industrial by-products for soil stabilization has captured extensive interests in recent years (Moon et al., 2013; Tsang et al., 2013). This can be regarded as “green remediation” in a broad sense, because such approach addresses the problems of contaminated soil/sediment while enabling resource recycling and mitigating the overall environmental impact (e.g., carbon footprint and landfill disposal) associated with solid waste management (Tomasevic et al., 2013; Tsang and Yip, 2014). Therefore, this study proposed to utilize different types of selected waste materials as supplementary binders

to facilitate cement hydration and as coarse aggregate to strengthen the matrix structure of stabilized/solidified sediment, such that the remediation process could be low-cost and low-impact while creating new reuse options.

It was suggested that the mechanical strength of stabilization/solidification could be improved via the pozzolanic reactions and pore filling in the structure by the addition of fly ash (Zentar et al., 2012; Tomasevic et al., 2013; Tsang et al., 2014) and/or ground seashells (Lertwattanaruk et al., 2012; Moon et al., 2013). The fly ash contained high contents of glassy silica and alumina, whereas ground seashells were calcium carbonate-rich kitchen waste, thus they could assist the formation of cementitious compounds. On the other hand, it was demonstrated in the construction industry that natural aggregate could be substituted by bottom ash from coal combustion power plant (Siddique, 2010; Arenas et al., 2011), bottom ash from municipal solid waste incinerator (Dabo et al., 2009; Gines et al., 2009), cullet from beverage glass bottles and recycled aggregate from construction waste (Kwan et al., 2012; Kou and Poon, 2013). These widely available waste materials were found to be relatively hard and inert, rendering them potentially suitable as coarse aggregate to improve the stabilized/solidified sediment for wider applications in sustainable urban development.

To assess the recycling applicability of the contaminated sediment, uniaxial compressive strength of the stabilized/solidified sediment blocks was measured and compared with the ASTM Standards. The mixture design was optimized for maximizing the potential of waste recycling and the mechanical performance of the sediment blocks. The crystalline mineral phase was analyzed by using X-ray diffraction (XRD). The microstructure and pore size distribution were investigated by using scanning electron microscopy with energy dispersive X-ray analysis (SEM-EDX) and mercury intrusion porosimetry (MIP). The toxicity characteristic leaching procedure (TCLP) was used to evaluate the acceptance criteria for reuse in terms of metal leachability.

2. Experimental methods

2.1. Contaminated sediment and waste materials

Contaminated sediment was dredged from the top 0.5 m of the To Kwa Wan Typhoon Shelter in Hong Kong. The total metal concentrations (average) in the sediment were 510 mg kg⁻¹ of Cu, 250 mg kg⁻¹ of Zn, 100 mg kg⁻¹ of Pb, 82 mg kg⁻¹ of Cr, 29 mg kg⁻¹ of Ni, and 0.8 mg kg⁻¹ of Cd, due to discharges from industrial areas in San Po Kong and former Kai Tak Airport in the past (HK EPD, 2013). According to the management guideline in Hong Kong, the sediment was considered highly contaminated (Category H) because the metal concentrations exceeded the Upper Chemical Exceedance Levels (HK EWTB, 2002). The dredged sediment was found to contain 68.9% moisture content, 34.6 g kg⁻¹ salinity (WTW inoLab Cond 720), and 6.4% organic matter (loss on ignition at 550 °C). The sediment sample was oven-dried, crushed to pass through 2.36-mm sieve, and re-saturated to a moisture content of 60% (i.e., same as the water-to-binder ratio, which was determined for optimum workability) right before subsequent recycling experiments. The sediment sample was composed of 6.1% sand, 74.8% silt, and 19.1% clay size fractions (on a dry mass basis, determined by wet sieving and hydrometer tests), thus it was recycled as fine aggregate.

The primary binder used in this study was ASTM Type I Ordinary Portland Cement (OPC). Lime (99% CaO), coal fly ash (CFA, ASTM Class F) obtained from CLP Power HK Ltd, as well as Blue mussel shell (MS) and Canadian whelk shell (WS) from Hotel ICON in Hong Kong (oven-dried and ground to powder size) were stud-

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